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Characterisation and Suitability of a CdTe Detector for Strontium 90 Assay in Groundwater

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Abstract

This presents a design of an in-situ detector for monitoring strontium 90 activity in groundwater at nuclear decommissioning sites. Current techniques for monitoring strontium 90 activity are lab-based and are time consuming, expensive and produce secondary waste. To alleviate these problems, a proof-of-concept detector has been developed which will be deployed in-situ, directly into groundwater boreholes. A compact detector was designed, housed in a waterproof casing and its initial performance as a strontium 90 detector in water was examined. A 10 x 10 x 1 mm cadmium telluride detector was paired with a appropriate charge sensitive amplifiers and readout systems to make a 118 x 83 x 40 mm sensor which was submersed in water. A 35.79 MBq collimated strontium 90 source was used to determine that the sensor was capable of counting beta particles from a range of 49.50 mm in water. The detector was held at a reverse bias of 80 V for 290 minutes, the average counts observed in 5 minute intervals went from $1.32 \times 10^6 \pm 1.14 \times 10^3$ to $1.28 \times 10^6 \pm 1.13 \times 10^3$. However, the results presented have established a viable proof-of-concept for an in-situ detector for strontium 90 assay in groundwater.

Key words: strontium, beta, radiation, detection, photodiode,

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²All colour images should be black and white for print

1. Introduction

Strontium 90, ^{90}Sr , is a by-product of nuclear fission and it is one of the prominent beta emitters found in nuclear waste at decommissioning sites such as Sellafield. It has a half-life of 28.8 years and decays via beta decay, releasing particles with energies up to 0.546 MeV. Its daughter yttrium 90, ^{90}Y , has a half-life of 64 hours and its decay releases beta particles with an end-point energy of 2.278 MeV [1]. ^{90}Sr shares similar chemical properties with calcium and can accumulate in calcium rich areas of the human body, such as bone structures, where irradiation can induce leukaemia and other bone cancers [2]. Contamination of groundwater at decommissioning sites is typically the result leaks from storage tanks, separation areas and ponds associated with fuel and waste storage. Fig. 1 depicts the scenario in which the detector will be deployed. ^{90}Sr which enters the ground may disperse and enter the groundwater table forming mobile plumes. At Sellafield Sr-90/Y-90 is the most common strong beta emitter in contaminated groundwater and greatly exceeds the WHO drinking water guidelines of 10 Bq/L [3] in many boreholes. While these waters are not exploited for drinking water purposes there is a long-term requirement to monitor the distribution and behaviour of this contamination on site.

Routine monitoring of ^{90}Sr is an expensive and time consuming process which produces secondary waste through collection of samples and laboratory analysis. Current techniques require manual sampling from underground boreholes. Tests often take months to produce results, as groundwaters are stored for weeks to allow for ^{90}Y ingrowth, and many of the techniques require the use of hazardous solvents and acids [4].

This research is investigating the feasibility of a cadmium telluride (CdTe) in-situ detector for the assay of ^{90}Sr in groundwater. CdTe is a compound semiconductor material which has become popularised and it has many properties that make it a suitable material for the detection of ionising radiation. CdTe has a high atomic number, 50, which makes it an effective absorber of ionising radiation and a wide bandgap, 1.44 eV, allowing for room temperature operation. It has gained traction in the fields of medical physics and astrophysics where it has been used to develop X-ray and gamma ray detectors [5]. However an increased atomic number is also associated with

Figure 1: A depiction of the scenario in which the detector will be deployed. Waste from decommissioning sites leaks into the groundwater where it is monitored via boreholes.

a higher rate of backscattering at the detector surface and this limits the number of particles completely absorbed within the detector.

As the detector must operate in a compact environment, it was important to use a detection medium which is small, capable of stopping the radiation of interest and does not require extensive cooling or other additional support systems. For these reasons, room temperature semiconductors were considered an appropriate candidate. A typical semiconductor detector consists of three layers. An anode, a cathode and an intrinsic layer of charge carriers. When beta radiation passed through the intrinsic layer, it does so through a series of Coulombic interactions which separate electron-hole pairs. In an ideal detector, free from defects which inhibit charge collection through charge trapping and recombination, the number of charge carriers generated is proportional to the energy deposited by the radiation in the detector. As the detector operates under a reverse bias voltage the electrons and holes are swept towards the anode and cathode respectively. The resulting current pulse can be detected, and its magnitude is equivalent to the energy lost by the radiation in the detector.

Previous research has investigated the performance of detectors of varying dimensions and material composition for the detection of ^{90}Sr in groundwater [6]. CdTe and GaAs were identified as suitable materials for the task with

similar properties and potential performance.

One of the drawbacks associated with CdTe is known as polarisation. This time-dependent effect occurs after the biasing of the diode and results in the reduction photopeak amplitude, lessening counting rate and diminishing the energy resolution [7, 8]. Polarisation can occur in CdTe as a result of radiation flux incident on the detector or after the detector has been biased for some time [9]. This is a result of disruptions in the uniformity of electric field across the material stemming from alterations in charge trapping in deep level states [10]. Detectors polarised by the biasing voltage can recover after the applied voltage is removed for a short time [11]. The stability of CdTe detectors can otherwise be improved by increasing the biasing voltage or operating at low temperatures [7].

The detector will be deployed in boreholes at decommissioning sites, where it will monitor beta activity by being submersed directly into contaminated groundwater. Beta particles have a limited range in matter due to their Coulombic interaction with nearby atoms. Their path through matter is characterised by large deflections where significant fractions of their energy can be lost. This can be accompanied by the radiative Bremsstrahlung process, where low energy X-rays are emitted as the beta particle decelerates. Additionally, beta particles are emitted over a continuous energy spectrum. These properties have presented difficulties when designing beta detectors.

^{90}Sr is the predominant beta-emitter found at Sellafield and its activity tracks with total beta activity. Beta emitters can be split into two groups, strong and weak, depending on their emission energy. The weak category includes carbon-14, ^{14}C (0.156 MeV), technetium-99 ^{99}Tc (0.249 MeV), and tritium (0.019 MeV) [1]. It is expected that these emitters will be too weak to penetrate the protective layers of the detecting system in addition to traversing the groundwater [12]. The other chief beta emitter is caesium-137, ^{137}Cs , which decays via beta emission, up to 0.512 MeV, to barium-137m, $^{137\text{m}}\text{Ba}$, which releases a 0.662 MeV gamma photon as it relaxes to its ground state. ^{137}Cs is generally found in low concentrations at Sellafield [13] with only one monitoring point exceeding the WHO safe water limit of 10 BqL^{-1} . By eliminating weak betas from entering the detector and considering the scarcity of other strong beta emitters, it may be the case that ^{90}Sr will be the only significant beta emitter which the detector is both sensitive and exposed to. Some research has suggested it may be possible to use gross measurement of activity in ground water to identify individual radionuclides by linear regression [14]. The time for completion for existing methods of groundwater moni-

103 toring is typically 1-2 months. This detector is being designed to produce
 104 results on a much smaller timescale, from 1 hour to 1 day, thereby providing
 105 information on a daily basis. This has the benefit of allowing decommis-
 106 sioning sites to examine the data quickly to determine whether beta activity
 107 is present or not, and rapidly respond to unexpected spikes in groundwater
 108 activity resulting from new leaks. This could be used to compliment existing
 109 techniques and allow for a selective approach with existing techniques such
 110 as liquid scintillation counting for closer examination of ^{90}Sr activity [15].

101 However, successful application of these technologies to in-situ monitoring
 102 will result in a significant reduction in secondary waste, long term costs and
 103 will also provide results allowing more detailed understanding of the response
 104 of ^{90}Sr to hydro-geological and geochemical perturbations to be assessed.

105 2. Materials and Methods

106 Groundwater boreholes are up to 50 mm in diameter, and filled with wa-
 107 ter. Therefore a robust and compact detector must be designed to operate
 108 in this environment. The average annual groundwater temperature at Sell-
 109 afield in 1997 was 10.67 °C [16]. This may be significant as some research
 110 has found that cooling to the region of 0-10 °C may have improve detector
 111 stability and its performance over time when compared to room temperature
 112 operation [7].

113 2.1. CdTe Diode

114 After considering the availability of semiconductor detectors on the mar-
 115 ket, and the feasibility of semiconductor fabrication, a commercial 10 x 10
 116 x 1 mm CdTe semiconductor detector was selected (supplied by AcroRad,
 117 Japan). The CdTe detector is constructed in an Ohmic configuration with
 118 20 nm thick platinum anodes and cathodes sandwiching a 1 mm thick CdTe
 119 crystal. This structure was mounted to an alumina substrate with gold wire
 120 bonds connecting to conductive pins which protruded from the bottom of
 121 the mount. An additional pin was used to ground the mount, which aided
 122 in reducing the noise present in the detector.

123 2.2. Electronics

124 The complete detector system is comprised of a few essential components.
 125 The current pulse produced by the CdTe semiconductor is amplified and
 126 shaped so that is more easily read by the counting system. This is achieved

127 with a charge sensitive pre-amplifier (CSA), which acquires the signal from
 128 the detector while boosting the signal-to-noise ratio (SNR). The CSA sums
 129 the charge created within the detector on a feedback capacitor which releases
 130 an amplified current pulse, of amplitude V_o , from charge generated, Q_d , and
 131 capacitance C_f . This signal pulse is then shaped by a shaping amplifier, to
 132 produce a signal of Gaussian profile which is read by an analogue to digital
 133 converter (ADC) for signal processing. The CdTe sensor was paired with the
 134 α -CSA supplied by Micod with a capacitance, C_f of 1 pF and resistance,
 135 R_F , of 100 M Ω . This produced current pulses with a sensitivity of 36 mV per
 136 MeV when paired with the CdTe diode. The amplifier was supplied with 5
 137 V from a DC power supply.

Figure 2: A) Depicts a schematic of the components in the detecting system and their function. B) Displays the deployment of the detecting system underwater in the experimental set-up.

138 A CAEN (Italy) A7585D modular supply was mounted adjacent to the
 139 CSA. The CSA was connected to the high voltage output of the power supply
 140 and this was used to bias the attached semiconductor detector. The CAEN
 141 power supply was programmed to supply a consistent 80 V bias to the CdTe
 142 diode. This bias was selected in accordance with manufacturer guidelines for
 143 an Ohmic diode and helps to meet the performance and power requirements
 144 necessary for a small detector [17]. These components complete the sensitive
 145 part of the detecting system and the CdTe diode, amplifier and a CAEN
 146 power supply were housed in a 118 x 83 x 40 mm plastic box. Currently, this
 147 is larger than the required dimensions to fit into a borehole, however it would
 148 only take minor modifications to rearrange the components in a configuration
 149 which fits. A window was cut from the box above the CdTe sensor to reduce
 150 the attenuation of beta particles. The distance from the sensor to the window

151 was 0.7 mm and this gap was filled with air. The complete detector system
152 is illustrated in Fig. 2.

153 *2.3. Attenuation due to Waterproofing*

154 To make the detector submersible in water, it was wrapped in two layers
155 of low density polyethylene. This material was selected to ensure a water
156 tight seal around the detector while keeping the attenuation of beta particles
157 to a minimum by using a thin (0.027 mm) and low-density (0.91 g/cm³)
158 material [18]. The waterproof detector was attached to the bottom of a
159 plastic container with adhesive tape, which was filled with room temperature
160 tap water. The water, plastic box, and metal clamp holding the source were
161 all grounded. The output from the detector was recorded directly with the
162 MCA connected to a laptop.

163 *2.4. Data Acquisition*

164 A digital multichannel analyser from Avicenna Instruments (USA) was
165 used to collect the pulses from the CSA and count them. This device consists
166 of a fast analog-to-digital converter (ADC) which digitises pulses from the
167 CSA and passes them to a Field Programmable Gate Array (FPGA) for
168 digital pulse processing. An FPGA is an integrated circuit which contains
169 a matrix of logic blocks which are freely programmable by the user. In this
170 case, the FPGA was programmed to function as a digital pulse processor
171 to create a lightweight solution for a portable multichannel analyser. The
172 device consists of an initial analog conditioning stage for the signal acquired
173 from the detector, the conditioned signal is then digitised by a fast ADC and
174 a FPGA where the pulse is further processed and sorted into a histogram
175 in accordance with its amplitude. The ADC samples at 100 MHz and the
176 digital processing consists employs a trapezoidal filter [19] to categorise and
177 bin pulses according to their height. A peaking time of 5 μ s and flat top
178 time of 0.4 μ s were found to perform the best for the CdTe detector. This
179 was done in conjunction with software on a laptop, connected to the MCA
180 via USB cable.

181 *2.5. Source and Background Count*

182 A sealed and collimated ⁹⁰Sr source was used for these experiments. The
183 aperture diameter on the collimator was 1 mm, barrel length 3.5 mm and the
184 activity of the source at time of use was 35.79 MBq. When the source was
185 submerged in water, it was wrapped in a thin nitrile layer to prevent water

186 from entering the collimator and absorbing the radiation. There is uncer-
 187 tainty over the activity of the source before and after collimation. Therefore
 188 the experiment was designed in such a manner that the absolute activity of
 189 the source is not strictly relevant. Results in terms counts are framed in a
 190 relative sense. The question being asked is about the range over which the
 191 detector can be expected to identify ^{90}Sr decay above the background noise
 192 in the detector.

193 Experiments were carried out in the Nuclear Physics department at the
 194 University of Glasgow. A metal clamp was affixed to the plastic container
 195 and this was used to hold the collimated source above the detector, with
 196 the beam in alignment with the semiconductor detector. This clamp allowed
 197 for the source to be positioned at different positions and distances from the
 198 sensor. While the detector was in use, it was covered with a light-proof sheet
 199 to prevent any interference of optical photons while the lighting in the lab
 200 was turned on. Three background counts were taken, each of a duration of 5
 201 minutes, equivalent to the counting time in many experiments. The average
 202 background count recorded in the detector was 48 ± 7 counts, equivalent to
 203 0.16 counts per second. The background counts appeared to be randomly
 204 distributed, with all but two confined to the first 232 channels on the MCA.

205 *2.6. Deployment in Water*

206 The detector was deployed into a a water filled container, to mimic the
 207 real world use-case for this device. This allowed the likely operating range of
 208 the detector through water to be characterised. The detector was fixed to the
 209 bottom of a 10 litre plastic contained which was with room temperature tap
 210 water. This simulated the deployment of the detector in groundwater and the
 211 ^{90}Sr source was positioned directly above the window to the semiconductor
 212 detector. Initially, the source was in contact with the dual layers of low-
 213 density polyethylene (LDPE) across the window. The counts in the detector
 214 were recorded for five minutes. The source was then moved vertically to
 215 increase the distance from the window, allowing for a larger volume of water
 216 to fill the space between the sensor to the collimator.

217 The performance of the detecting system over a longer duration of time
 218 was investigated. As mentioned previously, CdTe detectors can suffer from
 219 a degradation in performance after being biased or exposed to radiation for
 220 a period of time [7]. To investigate this, the detector was biased to 80 V
 221 and this remained consistent for the duration of 290 minutes. The ^{90}Sr was
 222 clamped in a single position above the detector level with the LDPE window.

223 This meant the particles were transmitted through two layers of LDPE and
 224 a 0.7 cm air gap. The counts observed in the detector were recorded for five
 225 minutes at various intervals over the course of 290 minutes. The detector was
 226 covered with light-proof sheeting for the duration of the experiment while the
 227 ambient temperature of the room was held at a constant 20 °C.

228 3. Results

229 3.1. Attenuation due to Waterproofing

Figure 3: Counts observed in the detector after measurements at increasing distances in water. A linear least squares regression has been applied to 3 points of the graph to allow an estimate for the range over which the detector is effective, 55.86 ± 5.40 mm.

230 The ^{90}Sr source was clamped at a distance of 12 mm from the sur-
 231 face of the plastic container and hits were recorded for 5 minutes. Com-
 232 pared with the uncovered detector, there was 9.36% decrease in counts, from
 233 $1.27 \times 10^6 \pm 1.13 \times 10^3$ to $1.16 \times 10^6 \pm 1.07 \times 10^3$. This is accounted for
 234 by particles scattering and being absorbed by the plastic layers between the
 235 collimator and detector. This decrease in counts is sufficiently low given the
 236 statistics involved in this experiment, however this may limit the effective-
 237 ness of the device when it comes to counting low activity sources. However,
 238 improvement could be achieved by removing one layer of LDPE, or trialling
 239 different lightweight and waterproof materials.

240 3.2. Detector Performance In Water

241 The counts in the detector were recorded over five minute intervals and
 242 plotted in Fig. 3. The counting time was selected to ensure a sufficient
 243 number of counts were collected to reduce statistical uncertainty. When the
 244 source was positioned directly at the window, $5.2 \times 10^5 \pm 721$ counts were
 245 observed in the detector and this drops off to 767 ± 28 counts at 40.0 mm.
 246 This provides a count which is significantly greater than the background
 247 rate for the detector. Beta particle intensity may follow a linear relationship
 248 with absorber thickness as the counts diminish [20]. By performing a linear
 249 least squares fit to the data points in Fig. 3, it is estimated the
 250 detector is sensitive to beta particles from a maximum distance of $55.86 \pm$
 251 5.40 mm before the minimal detectable limit is reached. In this case, the
 252 minimal detectable limit, N_D , is defined as the minimum number of counts
 253 required to state with a 95% confidence level that radioactive decay has been
 254 detected above the presence of background noise, N_B , in the detector [21].
 255 This is calculated with the following equation:

$$N_D = 4.653\sigma_{N_B} + 2.706 \quad (1)$$

256 This demonstrates that it is still reasonable to detect beta particles at
 257 range and this will be important in real world scenarios. The larger the
 258 volume of water the detector is sensitive to, the greater counting statistics
 259 it can generate. However, it should be noted that this experiment does not
 260 closely replicate the real world deployment of the detector. The collimated
 261 beam is considerably different to the colloid of radionuclides expected to be
 262 found in groundwater. However, the results produced here can be used to
 263 inform future experiments. This information will be useful when designing
 264 future experiments with contaminated groundwater, providing a guide for the
 265 necessary volumes of water required and provides information for determining
 266 the sensitivity and efficiency of the detector.

267 3.3. Detector Performance Over Time

268 The results are plotted in Fig. 4. The mean number of counts recorded
 269 was $1.29 \times 10^6 \pm 1.13 \times 10^3$. The maximum number of counts, was the first
 270 measurement taken while the lowest $1.28 \times 10^6 \pm 1.13 \times 10^3$ was taken after
 271 290 minutes. This represents a 3.26% decrease in counts. It is clear from
 272 Fig. 4 that the counts decline at a rate of 0.026% per minute during the first
 273 120 minutes, and the counts observed in the remaining 180 minutes were

more steady only declining at a rate of $3.9 \times 10^{-4}\%$ per minute. The average counts over the first 120 minutes was 1.30×10^6 compared with 1.28×10^6 over the last 180 minutes.

The experiment was repeated but this time the reverse bias voltage applied to the detector was turned off between measurements. The results are plotted in Fig. 4, as a fraction of the maximum count recorded. The initial count sees the greatest number of hits in the detector, and this falls to 97.19% after 290 minutes. The mean number of counts observed in the detector as a fraction of the maximum count was 98.16%, which was greater than the average of 97.52% when the bias was applied between counts. A similar trend is seen here when both experiments are compared.

Figure 4: ^{90}Sr Counts recorded over 5 minute counting intervals during a 290 minute period, with the bias on and off between measurements.

Both experiments see a period rapid of decline, followed by relative stability. When the detector was unbiased between measurements, the number of counts reduced by approximately 0.021% per minute but this declined to a loss of 0.0023% per minute over the final 150 minutes of the experiment. This can be contrasted with when the bias was left on, there was a loss of 0.031% counts per minute initially, followed by a loss of $8.30 \times 10^{-4}\%$ per minute.

Removing the bias from the detector between counts produced results with a higher average count as a proportion of the initial count. The insta-

bility in detector performance may be due to a number of factors including
 deviations in operating bias and variations in temperature [7] and some re-
 searchers have investigated radiation damage as a potential factor [22]. The
 detector is housed next to the 80 V power supply which generates heat as
 it operates and has an internal temperature monitor. While the detector is
 not submerged in water and operating in a 20 °C room, the temperature in
 the detector case averages at 31.01 ± 0.05 °C. The power supply used to bias
 the detector was used to monitor the internal temperature of the detector
 cavity over a 120 minute period while submerged in water. It was found that
 the temperature within the case rose from 24.49 ± 0.05 °C to 26.62 ± 0.05
 °C . while the detector was submerged under water with a temperature of
 14.8 ± 0.05 °C . This heat produced creates an unstable environment for
 the detector [23], and may lead to variable detector performance until the
 temperature reaches equilibrium and detector performance stabilises. This
 instability in detector temperature may be interfering with counting per-
 formance over long periods of time. Turning the bias voltage off between
 measurements may have alleviated the build-up of heat within the detector
 case and allowed for a cooler and more stable environment for the detector.
 As noted [7], cooling the detector down to temperatures around 10 °C may
 produce beneficial effects when it comes to the consistency of detector per-
 formance over a long term operational period. As the groundwater at Sellafield
 has an average temperature of 10.67 °C [16], this may provide a suitable basis
 for some ambient cooling. Fluctuations in groundwater temperature should
 be accounted for in future development of the detector.

4. Conclusions

The detector presented has demonstrated its ability to detect the pres-
 ence of a ^{90}Sr source. The results presented have demonstrated an operating
 range of over 40.0 mm in water from a 35.79 MBq ^{90}Sr source as the beta par-
 ticles propagate through water. By performing a linear fit to the data points
 in Fig. 3, it is estimated that the range of beta particle for this detector in
 groundwater is approximately 55.86 ± 5.40 mm before the minimal detectable
 limit is reached. In the real world application, the source of beta particles
 will not be from a collimated beam, but randomly dispersed throughout the
 groundwater. Currently, this is difficult to replicate in a lab, but previous
 research [24] has indicated that this should be capable of detecting radiation
 within the interval of activities expected at nuclear decommissioning sites.

330 As the detector is being designed to be deployed in-situ and provide measure-
 331 ments of the groundwater activity, it is required to have stable performance
 332 over near real time. The detector was held at a reverse bias of 80 V for
 333 290 minutes, the average counts observed in 5 minute intervals went from
 334 $1.32 \times 10^6 \pm 1.14 \times 10^3$ to $1.28 \times 10^6 \pm 1.13 \times 10^3$. This discrepancy may
 335 become important when detecting activities closer to the lowest detectable
 336 limit. It is anticipated that this is due to temperature changes as the detector
 337 operates and produces heat. The deployment of CdTe sensors into ground-
 338 water boreholes must account for variations in environmental conditions and,
 339 given the sensitivity of the detector to temperature, must accommodate for
 340 fluctuations in groundwater temperature.

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344 6. References

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